Coherent Soft X-ray Scattering for Studying Nanoscale Materials

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Introduction and motivations

Reminders about x-ray coherence

X-ray resonant (magnetic) scattering

Speckle phenomena

Nanoscale complexity: examples and applications

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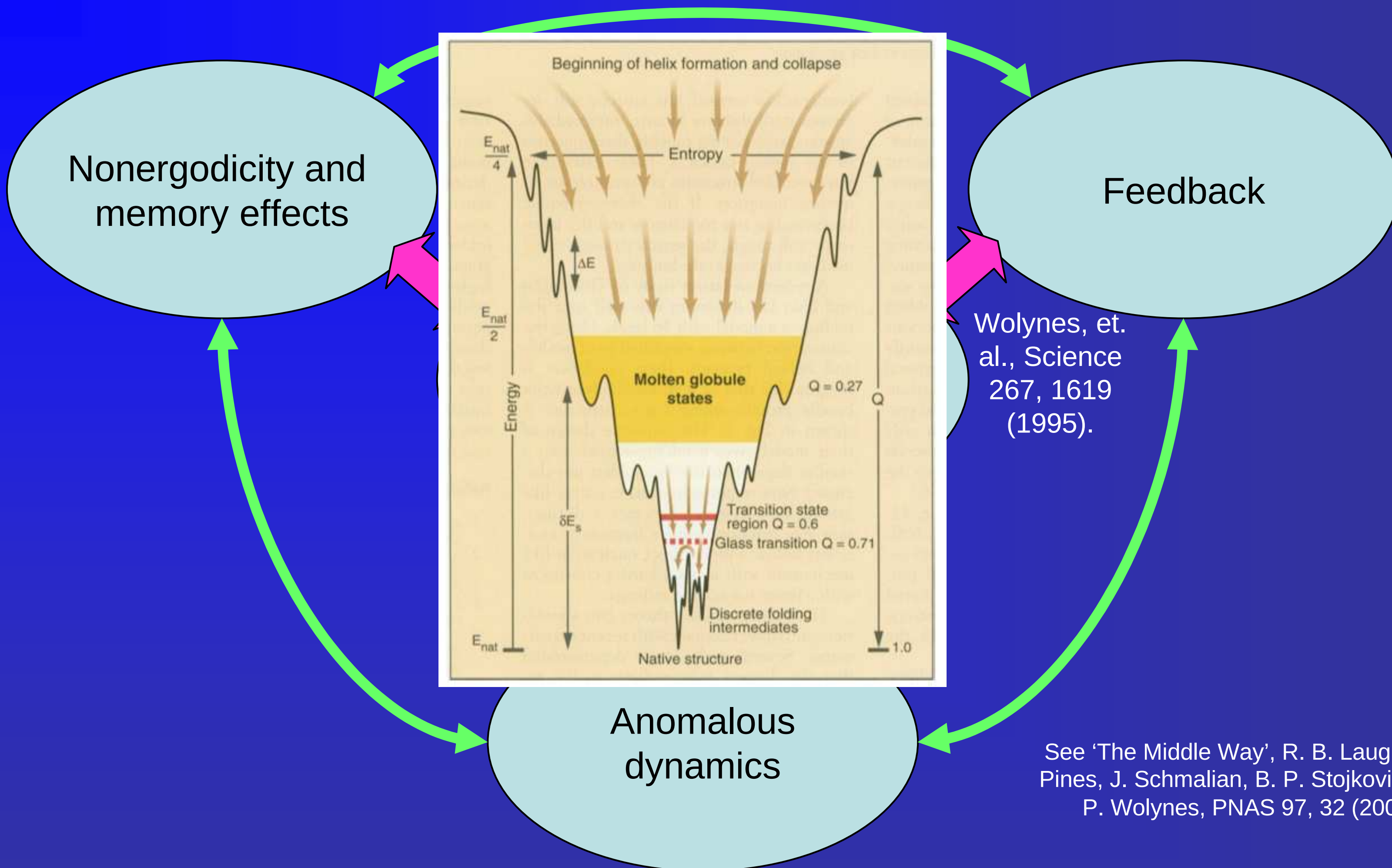
What Drives Material Complexity?

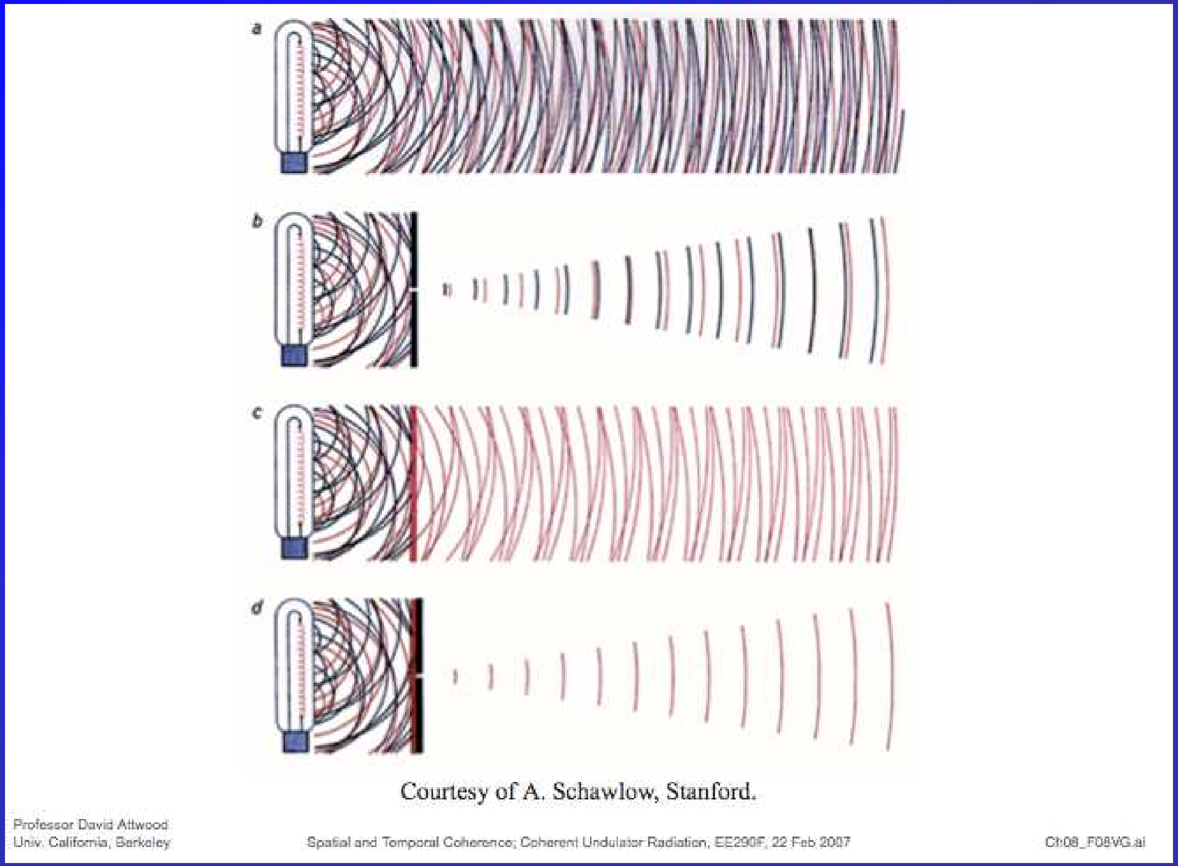


Wolynes, et. al., Science 267, 1619 (1995).

See ‘The Middle Way’, R. B. Laughlin, D. Pines, J. Schmalian, B. P. Stojkovic´i, and P. Wolynes, PNAS 97, 32 (2000).

These issues are often statistical in nature and can be usefully probed in terms of statistical averages, such as space-time correlation functions: S(q, t, T, H, E, j, . . .)

Some Reminders About Coherence

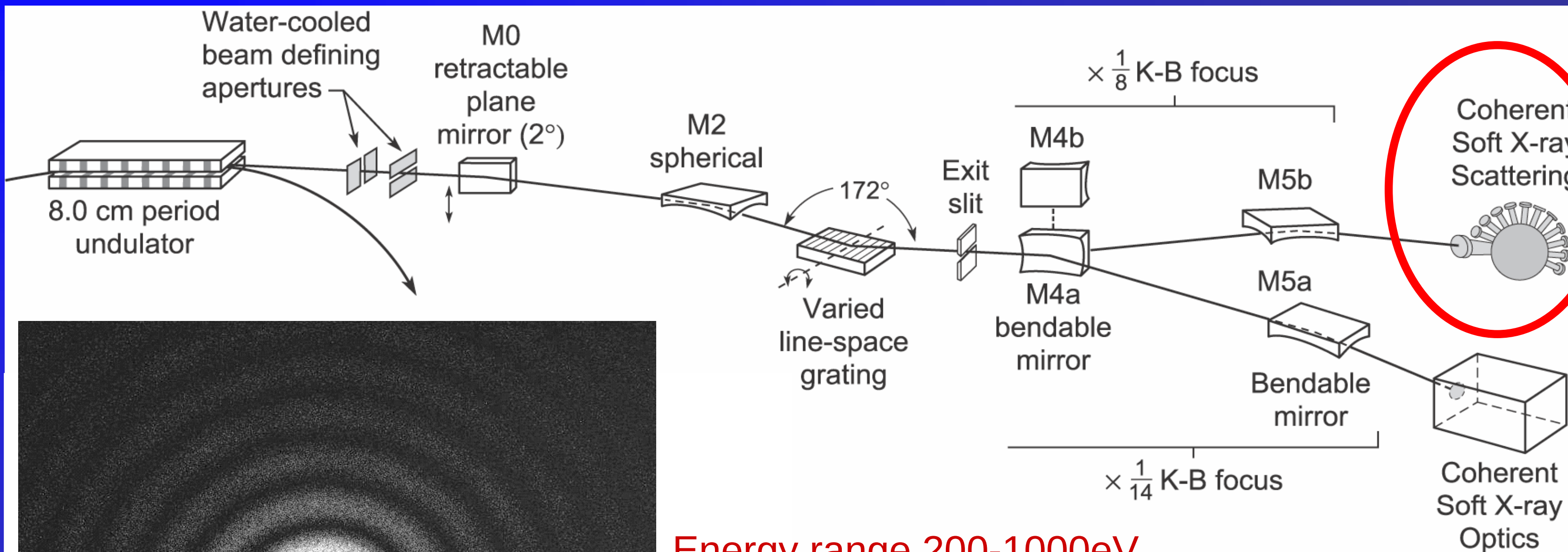


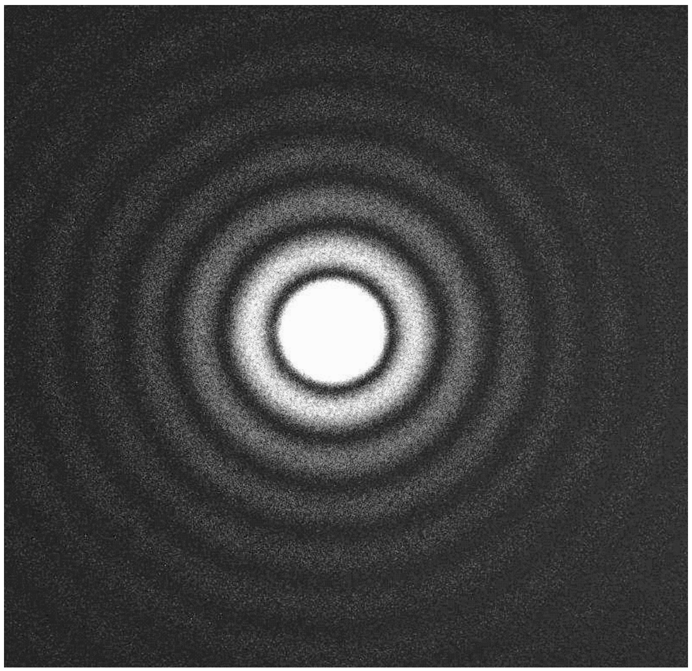
Extracting the coherent fraction from a partially coherent source:

$$F_{coh} = B \times (\lambda/2)^2 \times (\delta E/E) \times \text{beamline efficiency}$$

spectral brightness   transverse acceptance   longitudinal acceptance

ALS Coherent Soft X-ray Beamline (the current generation)





Energy range 200-1000eV

Moderate dispersion

8x demagnification of the source

Quality optics to preserve coherence

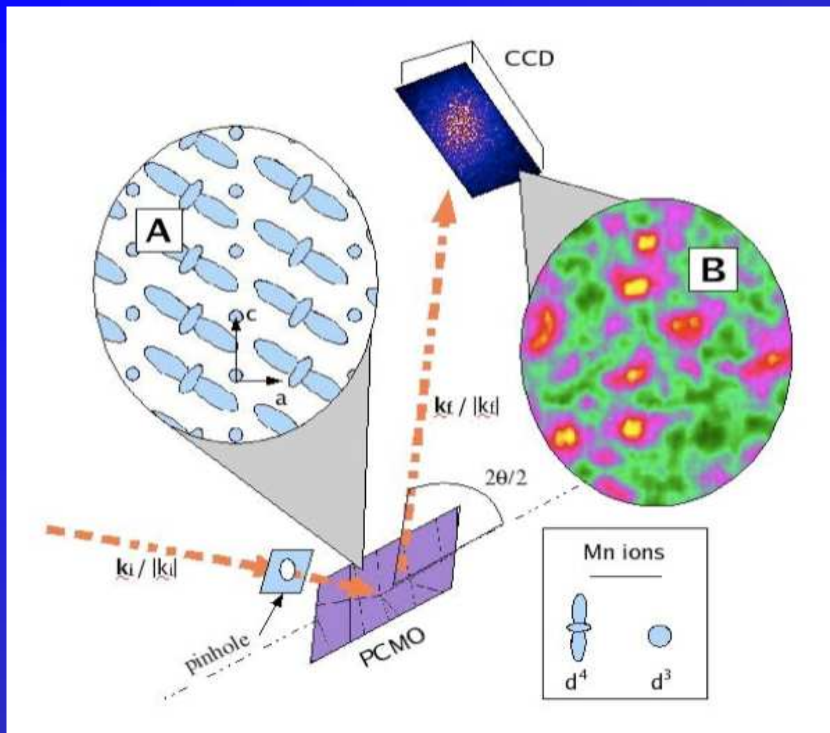
Coherent flux at 500eV: ~ 5×10^{10} ph/sec/0.1%BW

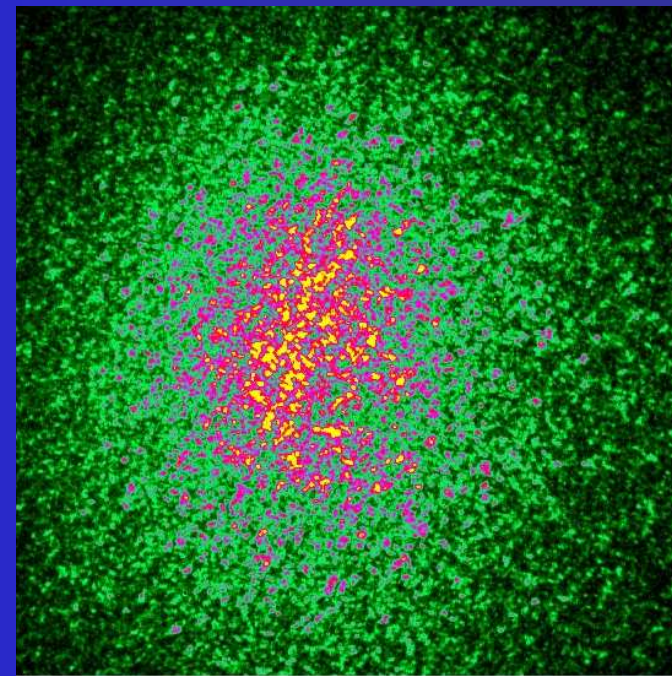
λ = 2.48 nm (500 eV)
d = 2.5 µm

Rosfjord et al. (2004)

Coh_Optics_BLwGraf.4.03.ai

'Imaging' Complexity with Coherent X-rays





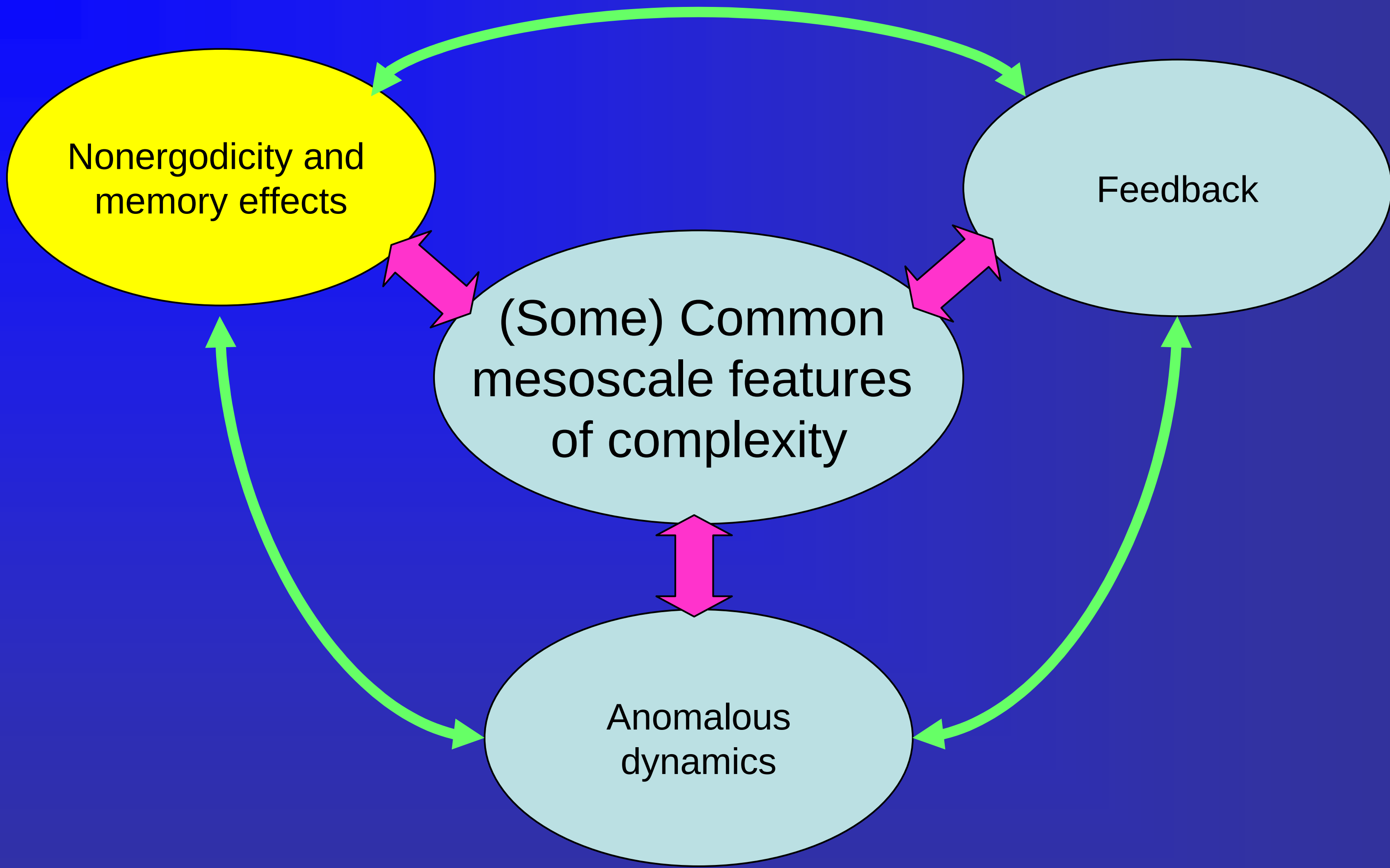
far

?

Lowest PCMO Bragg reflection at the Mn L_3 edge: the only way to image orbital domains?

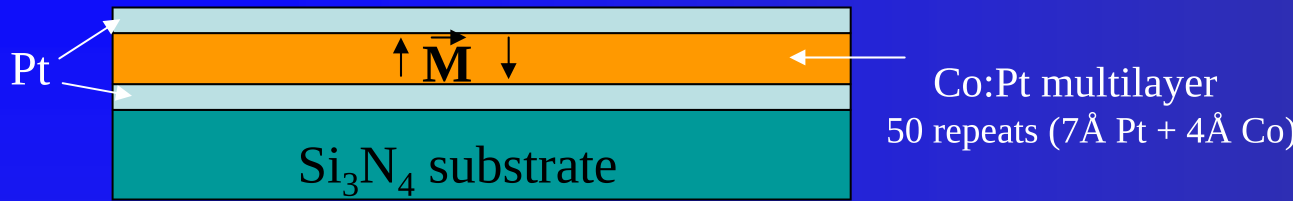
- Phase retrieval and imaging
- Speckle metrology, memory effects and external stresses, fields, currents
- Correlation spectroscopy and slow dynamics
- Feedback?

What Drives Material Complexity?

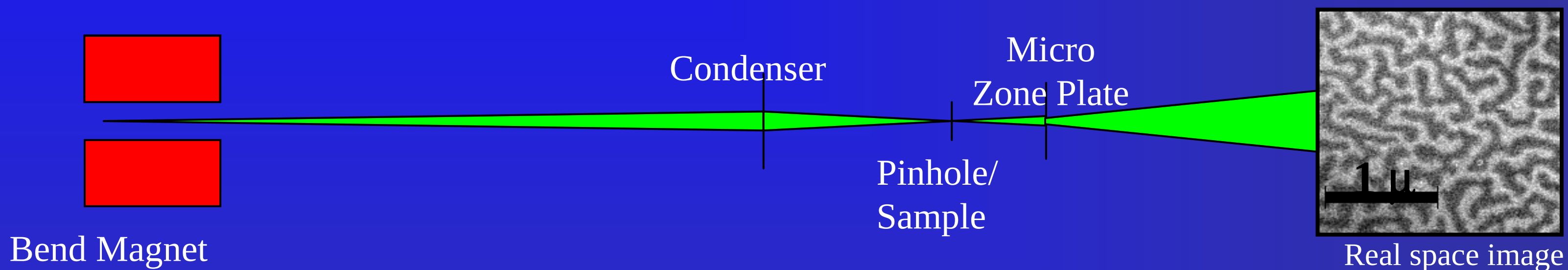


These issues are often statistical in nature and can be usefully probed in terms of statistical averages, such as space-time correlation functions: S(q, t, T, H, E, j, . . .)

Magnetic Domains in Real and k-Space



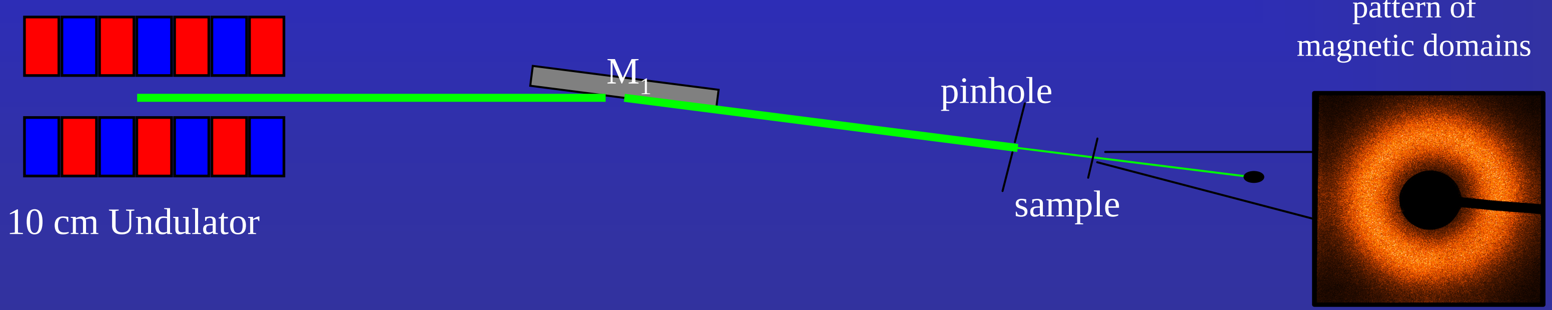
Magnetic contrast attained by operating near the Co L-edge



Real space image of magnetic domains

ALS XM-1 X-ray Microscope

Speckle-diffraction pattern of magnetic domains

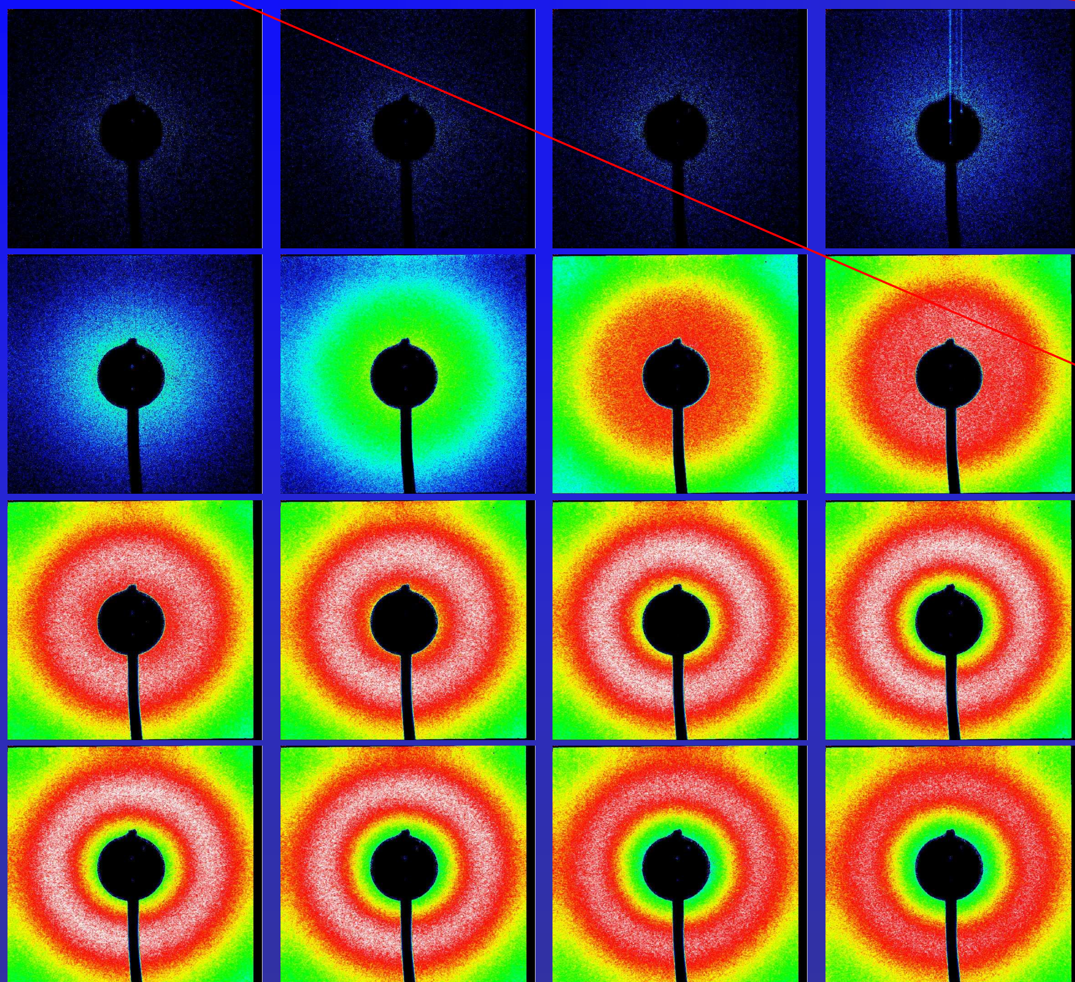


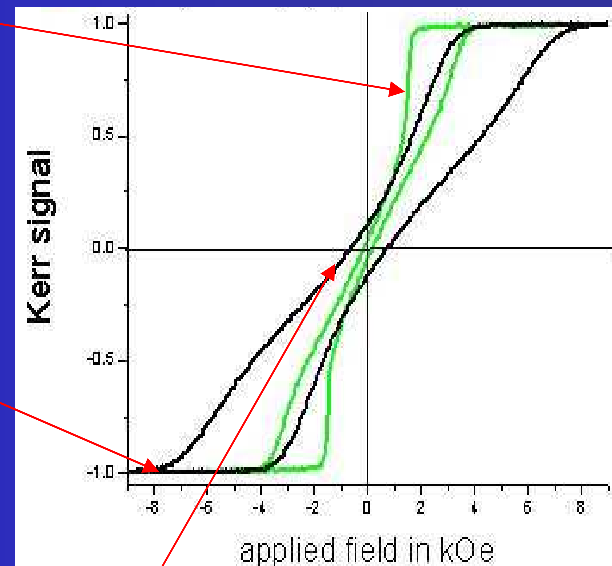
ALS BL7.0.1 (past) – BL9.0.1 ‘Blowtorch’ (recent) – BL12.0.2 CSX Beamline (current)

All Around the Magnetization Loop

saturation

nucleation

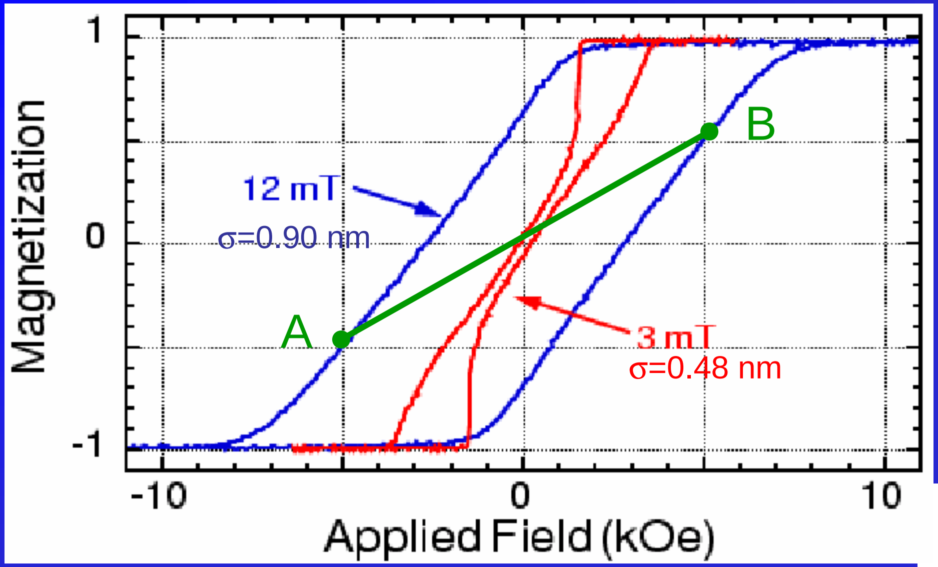




remanence

. . . . and on and on (Gb after Gb).

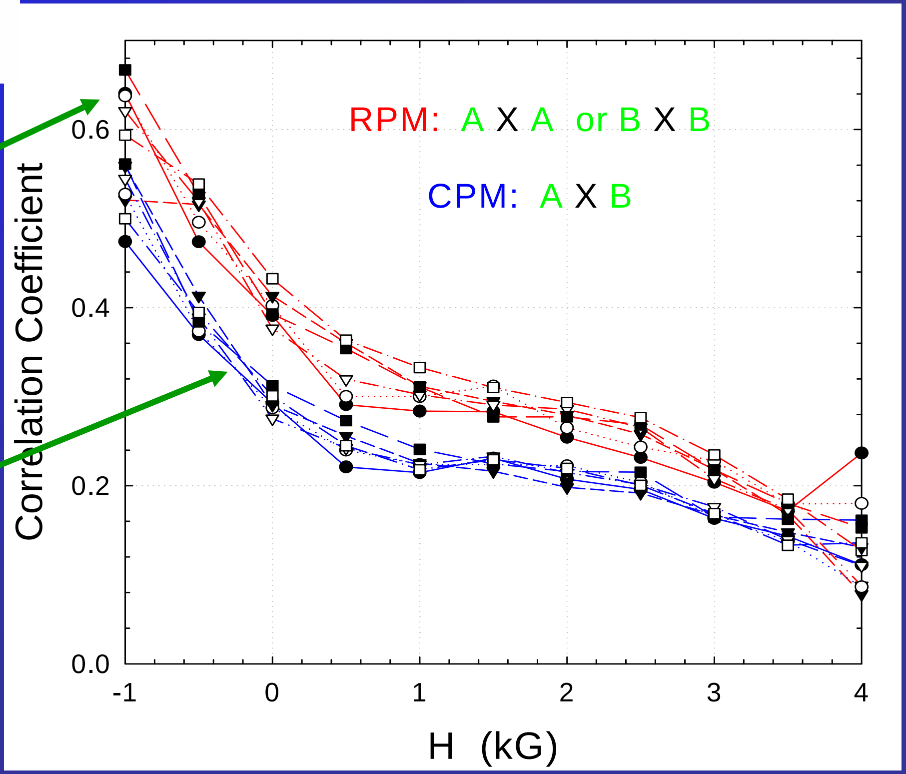
Microscopic Return and 'Conjugate' Point Memory



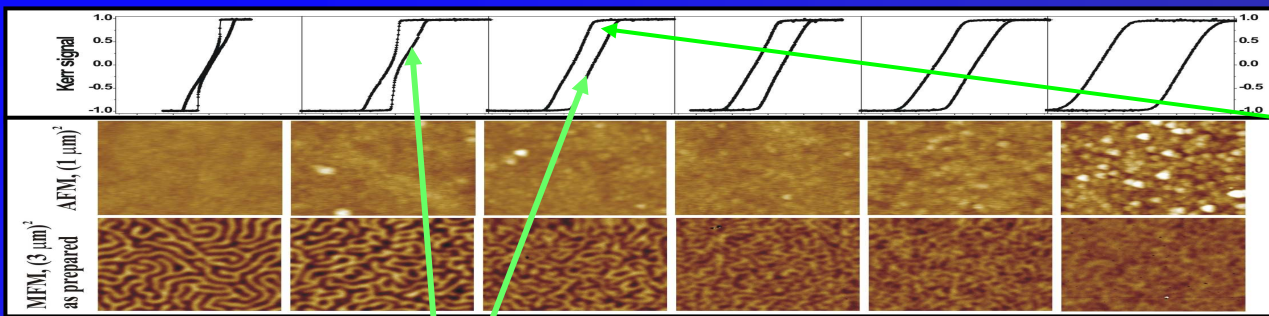
σ = 0.63 nm ('8.5 mT'):
Rougher films exhibit significant microscopic RPM and CPM, while smooth films do not.

Best memory near onset of reversal: first domains to nucleate have better memory.

Conjugate point memory is systematically ~20% lower than return point memory.



How (Dirty) Magnets Forget



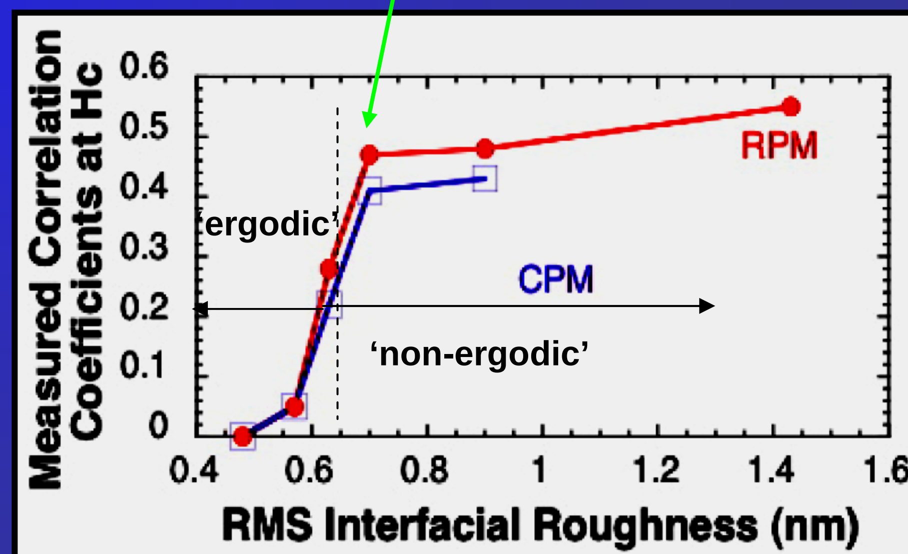
σ = 0.48 nm 0.57 nm 0.63 nm 0.70 nm 0.90 nm 1.43 nm

Roughness where a nucleation event disappears from the magnetization loop corresponds to an abrupt onset of RPM.

Theory of ‘crackling noise’ by Sethna* predicts an abrupt transition as a function of structural heterogeneity between a smooth magnetization loop and one with a distinct nucleation event, where a single Barkhausen cascade becomes macroscopic.

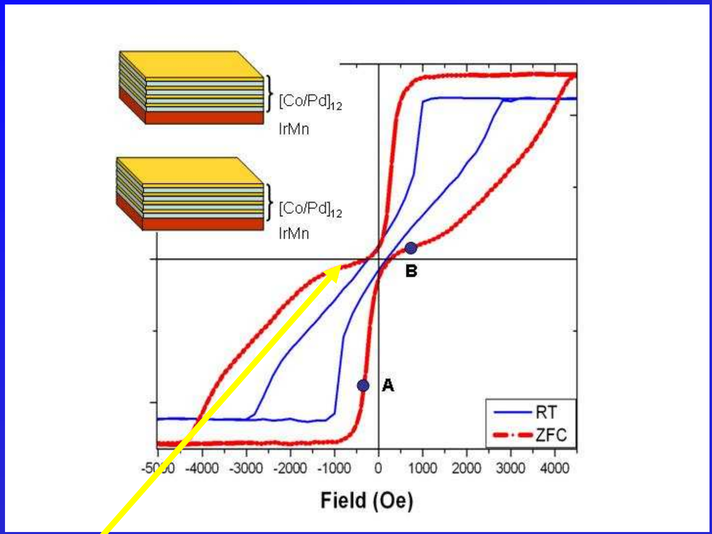
Multilayer perfection plays the role of a non-thermal parameter that allows us to control ergodic or nonergodic behavior.

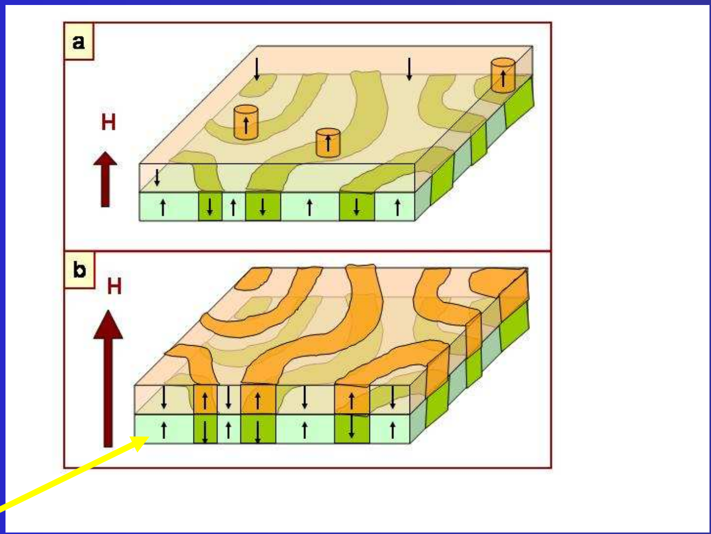
This T=0, random field Ising theory i) does not include dipolar interactions and thus does not predict measured loops very well, ii) predicts perfect return point memory, and iii) predicts zero complementary point memory.

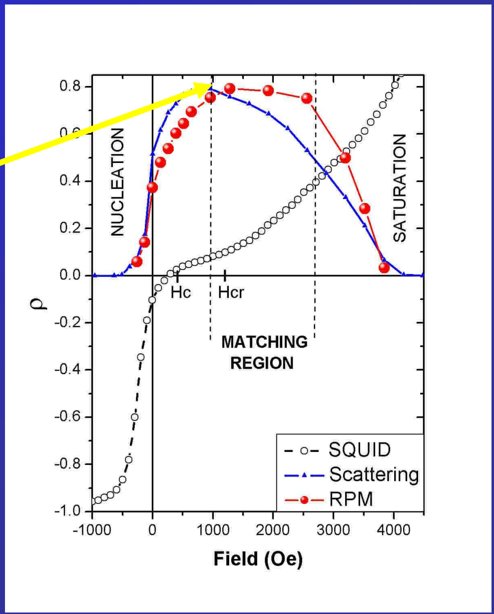


* see, for example, Sethna, Dahmen, and Myers, Nature **410**, 252 (2001).

Controlling Mesoscopic Memory with Exchange Bias Co:Pd - IrMn Films







‘Plateau’ in the magnetization loop after zero-field cooling caused by a ‘template’ of uncompensated spins in the AF layer which ensures good mesoscopic memory.

K. Chesnel, submitted.

Microscopic Memory: Future Issues and Avenues

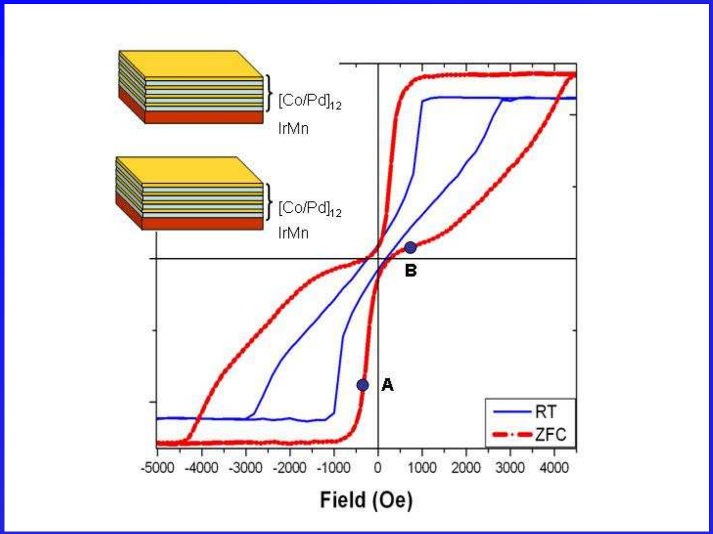
- Resolving the q-dependence of the correlation coefficient: interpolating between macroscopic and microscopic length scales

- More complex field protocols: easy vs. hard axis, rotation vs. inversion, memory in spring magnet systems

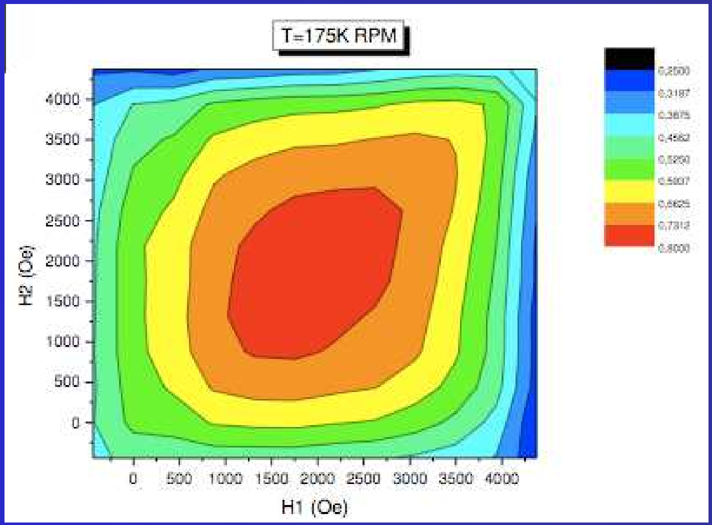
- High fields: microphase memory in complex oxides and the role of structural heterogeneity

- Correlation maps: a statistical probe of the funnel-shaped energy surface?

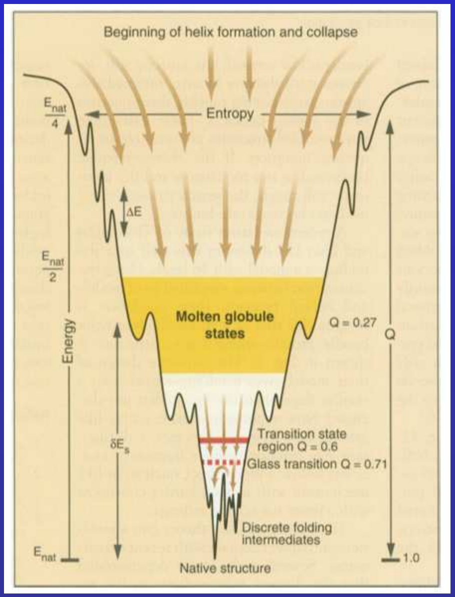
Memory Maps: *A statistical probe of the funnel?*



Macroscpic loop suggests microscopic memory



Full correlation map H_1 x H_2 delineates the region of high stability

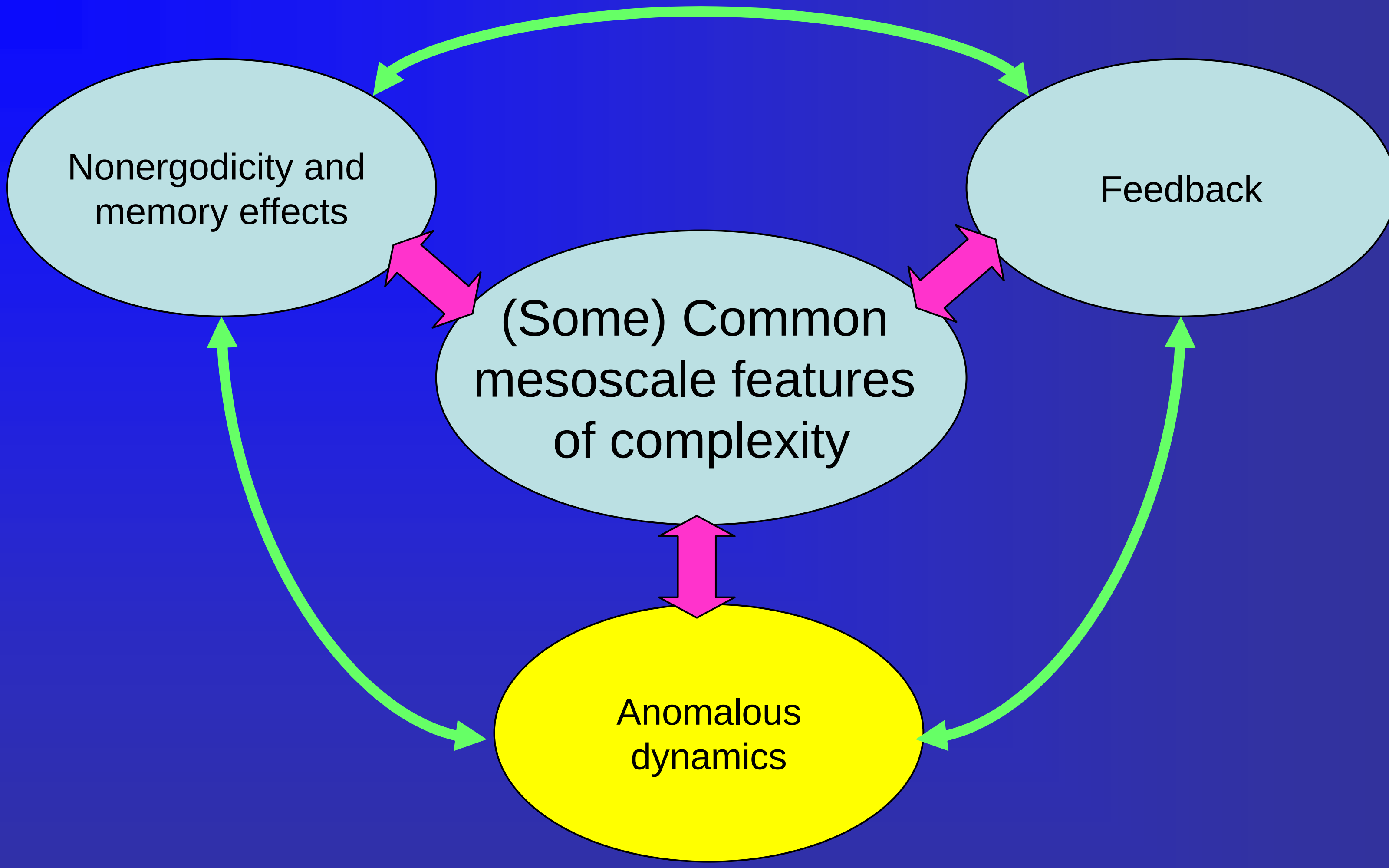


Isn't this roughly analogous to the configuration-space funnel suggested for protein folding?

Measuring the combined (q,T,H_{cool}) dependence of memory in exchange bias systems provides and excellent model for probing this relationship.

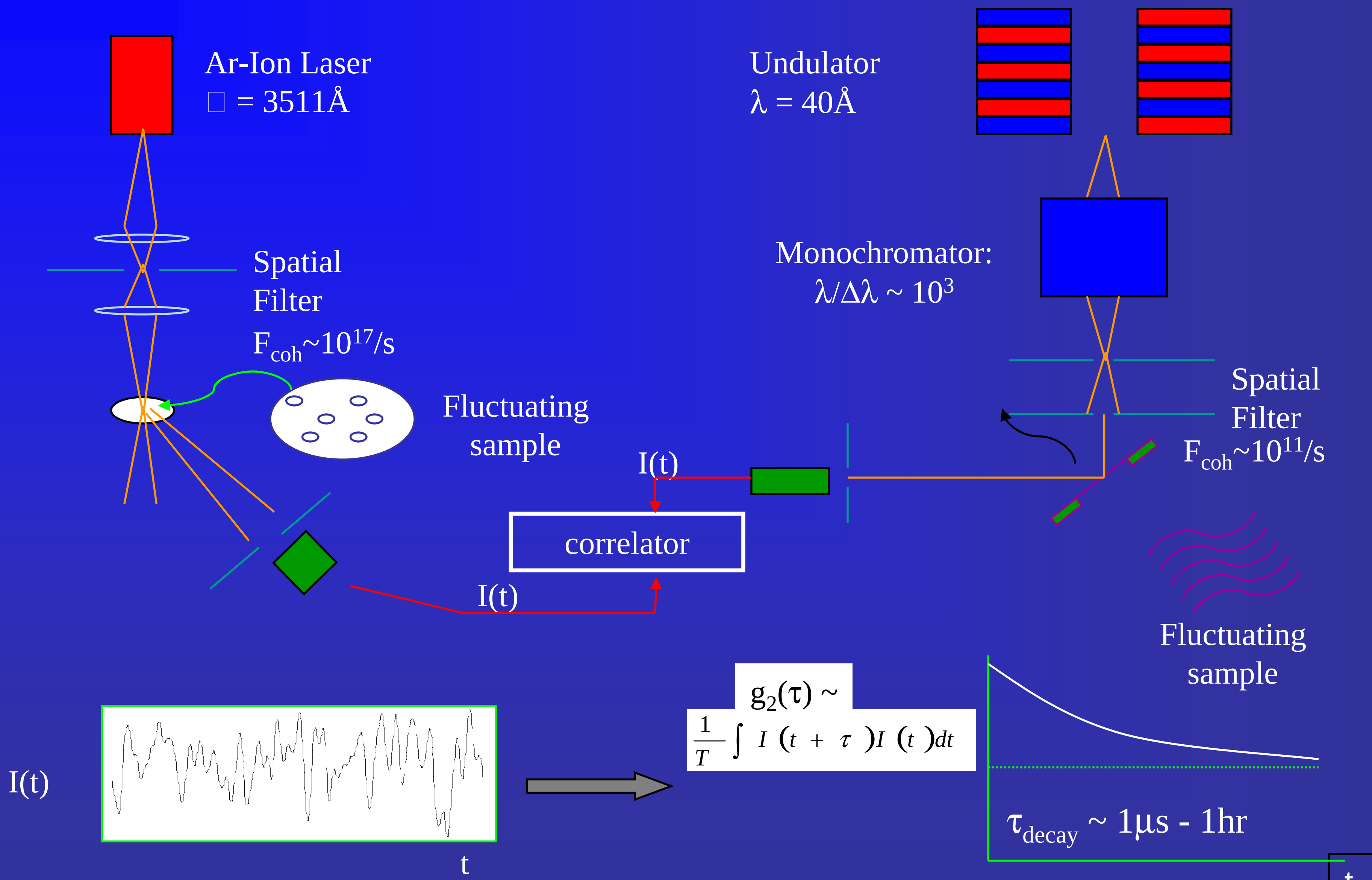
Physicists have been using magnetic systems as useful statistical model systems for some time . . .

What Drives Material Complexity?



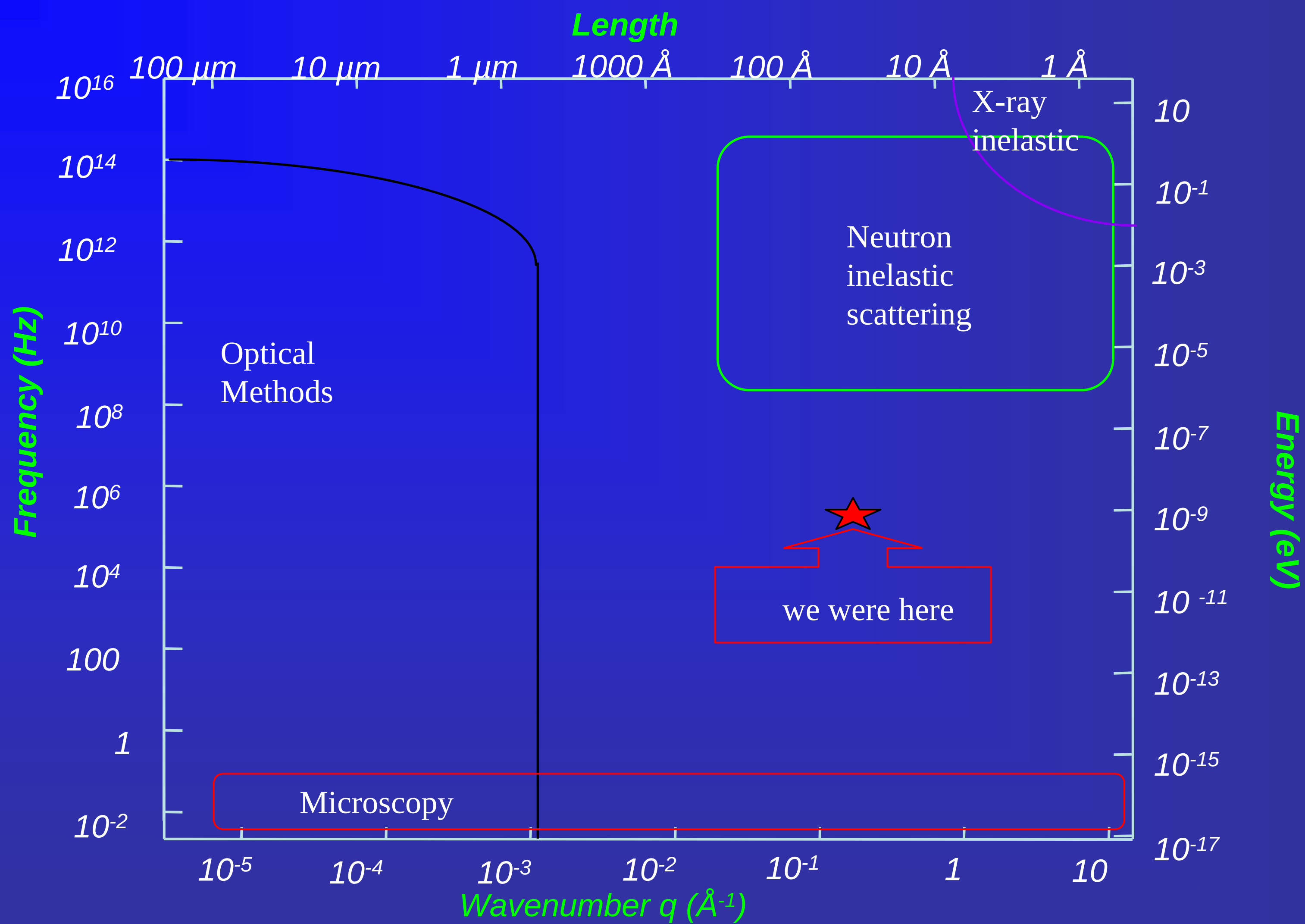
These issues are often statistical in nature and can be usefully probed in terms of statistical averages, such as space-time correlation functions: $S(q, t, T, H, E, j, \ldots)$

Dynamic Light Scattering



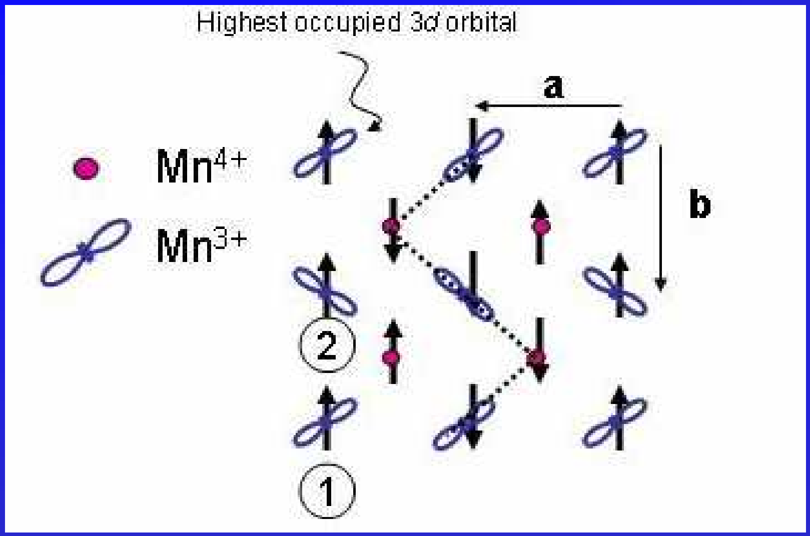
g_2(t) is the time Fourier transform of the dynamical stucture factor, S(q,ω).

Probing Hierarchies in Space and Time



★ "Soft X-ray Dynamic Light Scattering from Smectic A Films", A.C. Price, L.B. Sorensen, S.D. Kevan, J.J. Toner, A. Poniewrski, and R. Holyst, Phys. Rev. Lett., **82**, 755 (1999).

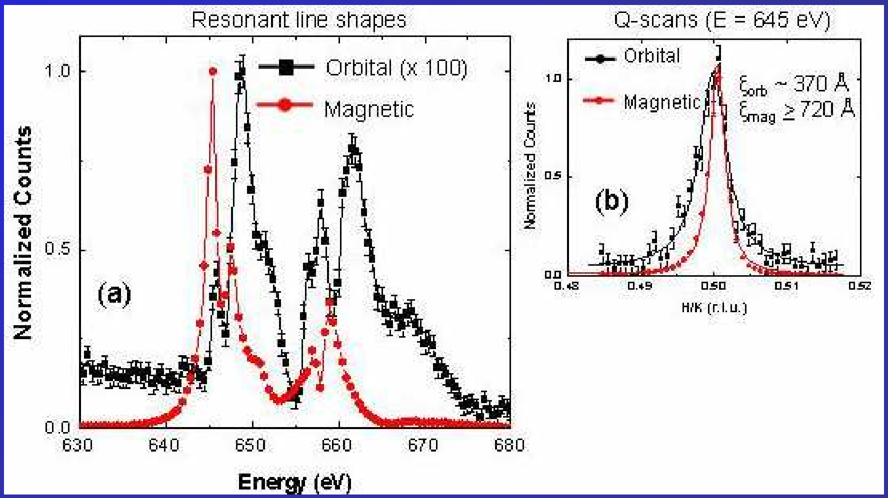
L-edge Structure in Orbital Ordered Manganites



‘Conventional’ picture of spin and charge ordering in $Pr_{0.5}Ca_{0.5}MnO_3$

- Mn 3d orbital physics helps determine the overall ground state;

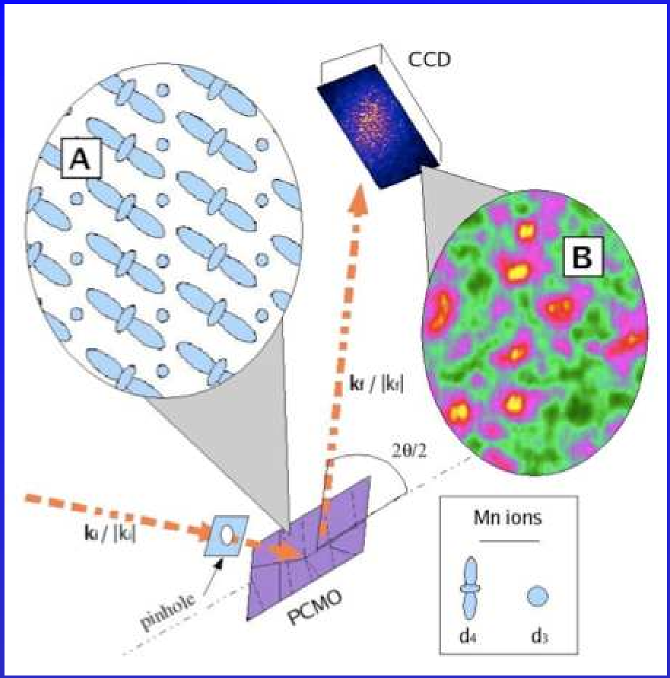
- L-edge anomalous diffraction offers a direct probe of how the atomic interactions couple to nanoscale spin and charge structures.



Resonant diffraction from magnetic- and charge-ordered superstructures (from X1B at the NSLS)

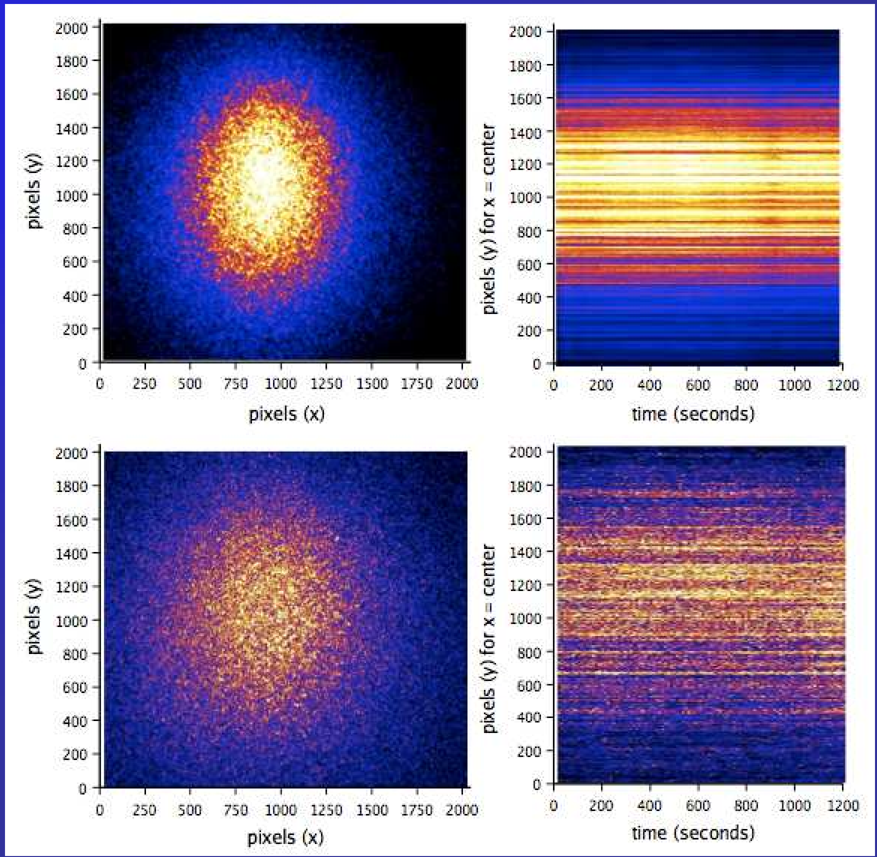
K.J. Thomas, J.P. Hill, S.Grenier, Y.-J. Kim, P. Abbamonte, L. Venema, A. Rusydi, Y. Tomioka, Y. Tokura, D.F. McMarrow, G. Sawatzky, and M. van Veenendaal, PRL 92, 237204 (2004).

How Does an Orbital Lattice Melt?

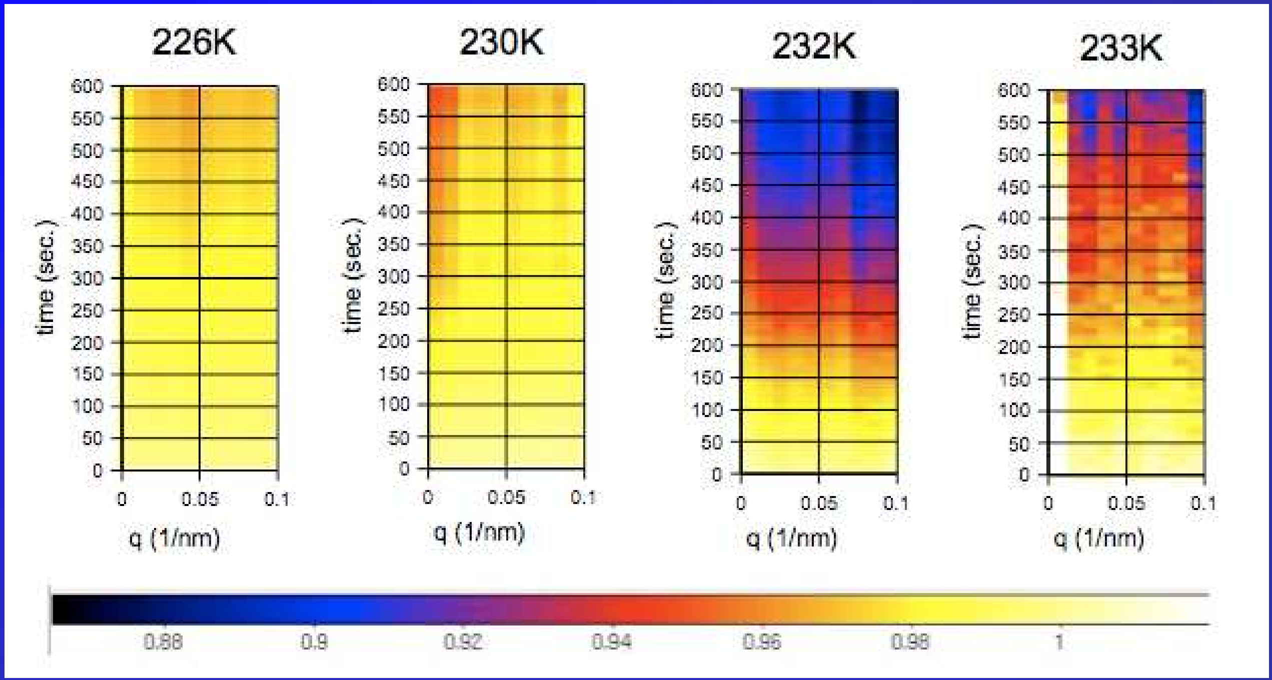


Schematic of scattering geometry sampling the (0,1/2,0) orbital-order Bragg peak that is broadened by finite-sized orbital domains.

Left: Images of the OO Bragg peak well below (top) and near the ordering transition. Right: Intensity vs time for a line of pixels through the middle of the Bragg peak indicating that the system remains mostly static even though the orbital peak broadens due to reduced OO correlation length.

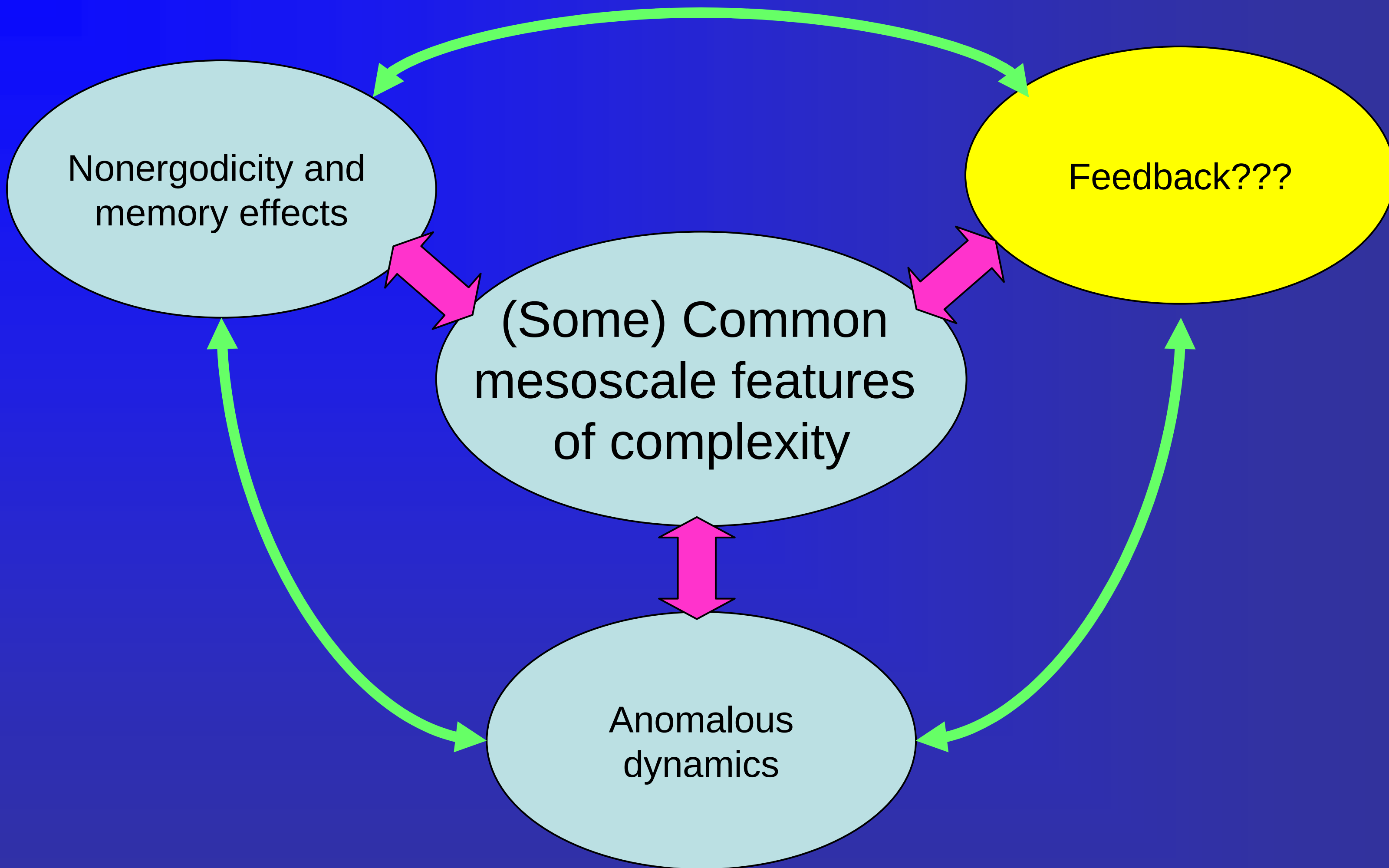


Orbital Domain Fluctuations: Ergodic and non-Ergodic Parts



- Orbital domains are essentially static below T ~ 232K, ρ ~ 1
- A small fluctuating component appears a few K below the OO/CO ordering temperature
- 'Frozen in disorder'? Orbital glass?

What Drives Material Complexity?



I believe that feedback, as very generally defined, plays a key role in both memory/nonergodicity and anomalous dynamics. . . but I think this is a target for various ultrafast studies, not for slower dynamics studied with correlation spectroscopy.

Conclusions: Coherence ➠ Correlations ➠ Complexity

Scattering coherent soft x-rays off complex materials maps their complexity into an easily-measured far-field speckle diffraction pattern with atomic, structural, and magnetic contrast.

These speckle patterns can be analyzed using various correlation function techniques to probe the microscopic memory and slow dynamics that are hallmarks of complexity.

Phase retreival and holographic imaging, in which such speckle patterns are inverted into real-space images, allow coherent x-rays to provide a unified view of real- and momentum-space - an important ingredient in probing mesoscle complexity (IMHO).